ENHANCEMENT OF MECHANICAL, THERMAL AND DIELECTRIC PROPERTIES OF HYBRID CARBON NANOTUBES AND ALUMINA IN EPOXY NANOCOMPOSITES

by

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Thesis submitted in fulfilment of the requirements
for the degree of
Master of Science

April 2015

ACKNOWLEDGEMENTS

I am grateful to Allah SWT with the permission and the gift; I was able to complete the thesis entitle Enhancement of Mechanical, Thermal and Dielectric Properties of Hybrid Carbon Nanotubes and Alumina in Epoxy Nanocomposites for the requirement of Degree Master of Science.

First and foremost, I would like to express my gratitude to my advisor Prof. Hazizan Md. Akil for his valuable guidance, support, and encouragements during my study at Universiti Sains Malaysia. His ingenious insights on research, efficiency and hard work had set an excellent example for me. I am fortunate to have him as an advisor & a mentor. Without his immense support, this dissertation would not have been possible.

I would also like to express my gratitude to my research group for their very helpful insights, comments and suggestions. All of you are not only excellent researchers but also fabulous person, and I really enjoyed working with you in such a friendly, encouraging, and active group. I wish all of them success in their careers. Moreover, I would like to thank the CREST (6050281) for sponsoring and giving financial assistance during this research work.

Finally, specials thank to my father and mother, Zakaria Bin Salleh and Hasnah Binti Darus, and my siblings for their love and inspiration. Thanks for believing in me when I did not believe in myself. This would not have been possible if were not for the love of my savior Allah SWT and his messenger Muhammad SAW. “Who is trying to be, he will succeed”

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LIST OF ABBREVIATIONS

ASTM	American Society for Testing and Materials
CNT	Carbon Nanotube
CTE	Coefficient of Thermal Expansion
CVD	Chemical Vapor Deposition
DGEBA	Diglycidyl Ether of Bisphenol-A
EDX	Energy Dispersive X-ray
FESEM	Field Emission Scanning Electron Microscope
HRTEM	High Resolution Transmission Electron Microscope
HYB	CNT-Alumina hybrid compound
MWCNT	Multi-walled Carbon Nanotube
MIX	CNT- Alumina physically mix
SWCNT	Single-walled Carbon Nanotube
UTS	Ultimate Tensile Strength
TMD	Trimethylhexamethylene Diamine
XRD	X-ray Diffraction

LIST OF SYMBOLS

%	Percentage
˚C	Degree Celsius
>	More Than
<	Less Than
GPa	Giga Pascal
MPa	Mega Pascal
GHz	Giga Hertz
KHz	Kilo Hertz
ppm/˚C	part per million/Degree Celsius
W/mK	Watt/meterKelvin
μm	Micrometer
nm	Nanometer
L	Length
Wt %	Weight Percent
mol	mole
Pa.s	Pascal-second
g/cm^3	Gram per cubic centimeter
g/l	Gram per litre

PENAMBAHBAIKAN SIFAT-SIFAT MEKANIK, TERMA DAN DIELEKTRIK BAGI HIBRID NANOTIUB KARBON DAN ALUMINA DALAM NANOKOMPOSIT EPOKSI

ABSTRAK

Penambahan nanotiub karbon (CNT) dalam nanokomposit polimer telah memberikan cabaran kepada penyelidik disebabkan oleh taburannya dalam matriks polimer. Kajian ini memberikan fokus terhadap sebatian hibrid CNT-alumina yang dihasilkan melalui pemendapan wap kimia (CVD) yang digunakan untuk mempertingkatkan taburan dan menambah baik sifat-sifat mekanik, terma dan dielektrik bagi nanokomposit epoksi. Sebatian hibrid CNT-alumina telah berjaya disintesis melalui kaedah CVD dengan menggunakan pemangkin nikel di bawah atmosfera metana pada suhu 800 °C. Bagi tujuan perbandingan, campuran CNT-alumina secara fizikal juga disediakan dengan menggunakan kaedah pengisaran bebola bagi tujuan perbandingan. Nanokomposit epoksi terisi sebatian hibrid CNT-alumina dan CNT-alumina yang dicampurkan secara fizikal telah dicirikan berdasarkan muatan pengisi (iaitu 1% - 5%). Sebatian hibrid CNT-alumina masing-masing mempunyai saiz antara 10 – 30 nm dan 12 % berat karbon berdasarkan analisis medan pancaran mikroskop imbasan electron pancaran medan (FESEM), mikroskop pemancaran elektron resolusi tinggi (HRTEM) dan serakan tenaga sinar-x (EDX), manakala analisis pembelauan sinar-x (XRD) mendedahkan kewujudan fasa karbon antara beberapa fasa alumina. Penilaian bagi sebatian hibrid CNT-alumina terisi nanokomposit epoksi menunjukkan sifat-sifat mekanik, terma dan dielektrik yang lebih tinggi berbanding CNT-alumina yang dicampurkan secara fizikal serta terisi nanokomposit epoksi. Peningkatan ini berkaitan dengan taburan seragam

sebatian hibrid CNT-alumina seperti yang diperhatikan daripada FESEM dan HRTEM. Penggunaan sebatian hibrid CNT-alumina terisi nanokomposit epoksi telah dibuktikan mampu untuk meningkatkan kekuatan tegangan sehingga 30%, modulus tegangan sebanyak 39%, kekuatan lenturan sebanyak 30%, modulus lenturan sebanyak 35%, kekerasan sebanyak 17%, konduktiviti terma sebanyak 20%, nilai suhu peralihan kaca sebanyak 25% dan pemalar dielektrik sebanyak 20% apabila dibandingkan dengan epoksi yang tulen.

ENHANCEMENT OF MECHANICAL, THERMAL AND DIELECTRIC PROPERTIES OF HYBRID CARBON NANOTUBES AND ALUMINA IN EPOXY NANOCOMPOSITES

ABSTRACT

The incorporation of carbon nanotube (CNT) in polymer nanocomposites has become challenges for researchers due to its dispersion in polymer matrix. This work focuses on CNT-alumina hybrid compound prepared via chemical vapor deposition (CVD) which is used to improve dispersion and enhance the mechanical, thermal and dielectric properties of epoxy nanocomposites. The CNT-alumina hybrid compound was successfully synthesized via CVD by using nickel catalyst under methane atmosphere at 800 °C. The physically mixed CNT-alumina was also prepared by ball milling method for comparison. The CNT-alumina hybrid compound and physically mixed CNT-alumina filled epoxy nanocomposites were characterized according to their filler loadings (i.e. 1% - 5%). The CNT-alumina hybrid compound had the size between 10 – 30 nm and 12 Wt % of carbon according to field emission scanning electron microscope (FESEM), high resolution transmission electron microscope (HRTEM) and energy dispersive x-ray (EDX) analysis respectively, while x-ray diffraction (XRD) revealed the existence of carbon phase among several phases of alumina. The CNT-alumina hybrid compound filled epoxy nanocomposites assessments showed higher mechanical, thermal and dielectric properties than the physically mixed CNT-alumina filled epoxy nanocomposites. This increase is associated with the homogeneous dispersion of CNT-alumina hybrid compound as observed from FESEM and HRTEM. It was demonstrated that the CNT-alumina hybrid compound filled epoxy nanocomposites are capable of increasing tensile

strength by up to 30%, giving tensile modulus of 39%, flexural strength of 30%, flexural modulus of 35%, hardness of 17%, thermal conductivity of 20%, glass transition temperature value of 25% and dielectric constant of 20% when compared to a neat epoxy.

CHAPTER 1

INTRODUCTION

1.1 Overview

The extensively growing area of nanoengineering materials will create many perspectives for polymer and composites determined by the final product of the polymer nanocomposites. In the late 1980s, polymer nanocomposites were commercially produced in both academic laboratories and research organisations (Komarneni, 1992). Roy (1992) and Komarneni et al. (1997) started to use the term ‘nanocomposites’ in 1984 to describe the fact that polymeric product contains two or more distinct materials in the nanoscale. Since that time, the term ‘nanocomposites’ has been globally recognized and described as a very large group of materials involving structures in the nanometer range between 1-100 nm (Gleiter, 1995). The properties of the nanocomposites have become an attraction to researchers due to size of the structures which is totally different from those of the bulk matrix. In 1988, the Toyota Group became the first company to utilize nanocomposites parts for their novel car models by using polymer/layered nanocomposites (Fukushima et al., 1988). Subsequently, numerous other companies also started to look into nanocomposites and this led to a dramatic development of research in this novel class of materials.

The reinforcement of polymer by using fillers such as inorganic or organic materials is typical in the production of modern polymer composites. Generally, conventional composites use micrometer particles, platelets or fibres as fillers in polymer composites. These conventional composites have been explored for many

years to be fully utilized in various industrial applications (Schadler, 2003). For examples, composites based on thermosetting resins are extensively used for structural materials product such as polymer, concrete, glass fibre reinforced epoxy, inorganic particle filled epoxy, etc. Usually, micro-filler particles are inactive and their main purpose is to reduce the cost of the final product. The most crucial factors that control the properties of polymer composites which contain inactive fillers are size, shape and distribution of the filler, while surface morphology and chemistry play a minor role. On the other hand, active fillers in polymer composites demonstrate a reinforcing effect of filler on mechanical properties depending on the morphology and interaction between filler and polymer matrix (Ajayan et al., 2006). Typically, polymer reinforced with micrometer active fillers exhibits improvement in hardness, however its tensile properties worsen due to stress concentration resulting from the presence of filler particle.

Polymer nanocomposites are developed by incorporating a small amount of nano-filler dispersed at a molecular level in the polymer matrix as a replacement for the conventional polymer composites. The homogenous dispersion of nano-filler particles provides very high surface area to volume ratio between filler and polymer matrix. A special characteristic of polymer nanocomposites is that a significant enhancement in properties can be achieved at low filler loading which consequently minimizes the weight of the final product (Mai and Yu, 2006).

Epoxy resins are a type of versatile polymer materials characterized by the existence of more than one epoxy group or oxirane ring located on their molecular structure. Similar to other thermosets, epoxy resin also forms a network on curing

with various types of curing agents. Nowadays, epoxy resin is recognized as one of the interesting class of polymers and used as a matrix for polymer composites due to its strong adhesion and excellent overall mechanical properties, including high strength and stiffness, high chemical, thermal, and dimensional stability, as well as good creep and solvent resistance (Hussain et al., 2006). The performance of epoxy resin can usually be improved by incorporating various type of including particulate filler and fibrous reinforcement. Recently, carbon nanotubes, as particulate filler in epoxy resin, have been widely used due to their remarkable improvements in mechanical, electrical, optical and thermal properties (Coleman et al., 2006a).

Over the last decades, carbon nanotubes have remained attractive to both engineers and scientists who are interested in their applications due to the unique combination of their properties (Meyyappan, 2004). Carbon nanotubes have progressed into one of the most intensively studied materials and they play a major role in co-triggering the polymer nanocomposites (Andrews and Weisenberger, 2004). Carbon nanotubes are regarded as seamless cylinders formed by wrapping graphene sheets with carbon atoms that are covalently bonded with each other through sp2 hybridization. Based on the number of layer of graphene sheets, they can be categorized as single-walled or multi-walled carbon nanotubes. These carbon nanotubes molecular systems are nanometer in diameter and up to centimeter in length which demonstrate length/diameter aspect ratio of more than 10^7 (Ebbesen, 1996). The contiguous carbon nanotubes possess a high degree of atomic-scale perfection and this makes them chemically inert as their close relationship with graphene. As for graphene under tension, the strength of the carbon nanotubes are two times higher than steel (Gojny et al., 2004). The melting point of the carbon

nanotubes in vacuum is approximately 3700 ˚C which is almost similar to graphite and greater than any metal (Jorio et al., 2007). Carbon nanotubes can act as ballistic conductors of electrons or behave as a semiconductor depending on the atomic structure and the diameter (Javey et al., 2003). In addition, carbon nanotubes are an excellent conductor of heat and reported to surpass the thermal conductivity of pure diamond (Jorio et al., 2007). Furthermore, carbon nanotubes also seem to be biocompatible in many environments (Smart et al., 2006).

In the meantime, a wide range of carbon nanotubes based hybrid materials have also been developed by the combination of carbon nanotubes with other materials. These hybrid materials with attractive features exhibit promising applications in the fields of nanobiotechnology, energy conversion, fuel storage, catalysis and electronic nanodevices (He et al., 2009). In particular, the micro supports could be either micrometric inorganic particles or various fibers. Instead of conventionally mixing two types of materials in a random way, carbon nanotubes are directly grown on the surface of the microscopic supports to form the nano-micro hybrid structures (Xie and Gao, 2007; Wang et. al., 2008; Jiang et al., 2007). The nano-micro hybrid structures have a reinforced interface between each carbon nanotubes and the micrometer support. At the same time, rather than in entangled state, carbon nanotubes could be distributed on the surface of micrometer particles. This is desired to avoid the agglomeration of carbon nanotubes and to obtain good dispersion in composites. Furthermore, hybrid materials in the composites favor to transfer efficiently the loads between the matrix and the fillers. Thus, enhanced properties of composites are expected to be achieved by using the hybrid materials as fillers in composites.

1.2 Problem Statement

The extraordinary properties of the CNT make it an ideal candidate to be used as multifunctional filler system in epoxy composites. Nevertheless, there are huge challenges for researchers to impart the intrinsic properties of the CNT due to the difficulty to disperse CNT in the epoxy matrix. CNT is usually present in the form of bundles or ropes due to the intrinsic van der Waals force attraction, its high surface area and its high aspect ratio (HowardáEbron, 2004). These characteristic of CNT can easily result in poor dispersion and significance level of agglomeration. Poor dispersion of the CNT will significantly reduce their reinforcement efficiency due to the CNT slipping each other when load is applied which will result in the formation of micro-voids in the epoxy composites.

Many researches have been conducted to overcome the above mentioned challenges i.e; to improve the dispersion of CNT in epoxy matrix (Du et al., 2007). Most of the techniques are dealing with processing issues rather than looking at the filler design aspect or architecture. The research includes investigation of different methods such as chemical functionalization (Yang et al., 2009), and physical blending (ultrasonication or high shear mixing) (Park et al., 2002). Chemical functionalization focuses on the surface treatment to the CNT structure to improve their chemical interaction with polymer matrix, and it has enhanced the dispersion of CNT. However, chemical functionalization method often causes structural defect that diminishes the intrinsic properties of CNT (Park et al., 2002). Hybridization of CNT with an inorganic filler is implemented to improve dispersion of CNT without damaging its structure. Nevertheless, conventional hybridization method by milling requires longer time which may damage the CNT structure. Furthermore, the